PATENT ABSTRACTS OF JAPAN

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(54) MANUFACTURE OF SILICONE COMPOSITE BODY

(57)Abstract:

PURPOSE: To easily manufacture silicone composite body consisting of surface protective layer and stress relaxation layer by a method wherein layer made of silicone composition, which turns into the form of gel by hardening, and silicone composition, which is provided on the surface of the layer and turns into the form of elastomer or the like, are hardened simultaneously.

CONSTITUTION: The silicone composite body concerned consists of base layer made of silicone gel and surface layer, which is formed on the surface of the base layer and made of silicone elastomer or silicone resin. In this case, at the manufacturing of the silicone composite body, firstly, layer made of hardening silicone composition, which hardens into gel-like hardened matter, is provided. Next, on the surface of the above-mentioned layer, layer made of hardening silicone composition, which is non-compatible with the above-mentioned composition and, at the same time, hardens into elastomer-like or resin-like hardened matter. In succession, the respective layers made of the above- mentioned compositions are hardened simultaneously. Thus, silicone composite body consisting of surface layer having high hardness and the like and base layer having low hardness and the like is easily manufactured by single hardening treatment.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application]This invention relates to the manufacturing method of the silicone complex which consists of base phase which consists of silicon gels, and a surface layer which consists of the elastomer or resin formed on this phase.

[0002]

[Description of the Prior Art]Since the gel hardened material of silicone is called what is called silicon gel and it has the outstanding characteristics, such as the stability of electric insulation and an electrical property, and pliability, As the electrical and electric equipment, potting of electronic parts, and an object for closure, especially, circuit elements for control, such as a power transistor, IC, and a capacitor, are covered, and it is used as covering material for thermal and protecting from a mechanical obstacle.

[0003]However, since the surface of silicon gel has strong adhesiveness, garbage and dust adhere easily and

there is a problem on the work of a molded product ******ing and not being made on silicon gel. When removing the garbage etc. which adhered since the mechanical strength was insufficient, there is a problem on which silicon gel suffers a loss. Methods of canceling a fault which was mentioned above include the method of providing a surface protection layer in the silicon gel surface so that it may state below. [0004](1) How to provide the protective layer of other organic system high hardness resin on the surface of silicon gel, [0005](2) How to carry out heat cure and to provide the silicone layer of the shape of an elastomer, or the shape of resin in the surface after giving the ORGANO hydrogen polysiloxane etc. on the surface of silicon gel and diffusing a gel surface as indicated to JP,1-25704,B, [0006](3) As indicated to JP,61-277414,A, The ORGANO hydrogen polysiloxane is given to the surface of the silicone gel composition of an uncured state, How to carry out simultaneous hardening of the surface layer from which it heated and an inside silicone gel composition and the ORGANO hydrogen polysiloxane were incorporated before being spread, and to provide a silicone elastomer layer on the surface of a silicone gel composition, [0007](4) The method of giving the constituent which becomes a silicone elastomer layer or a silicone resin layer by hardening to the surface of the hardened silicon gel, making it harden it, and providing a surface layer in it, etc. are known.

[Problem(s) to be Solved by the Invention] However, in the method of the aforementioned (1), since layer indirect arrival power is insufficient, it is easy to generate interlaminar peeling. So that it may not remain in the method of (2), not having hardened the ORGANO hydrogen polysiloxane on the silicon gel surface. The kind

of ORGANO hydrogen polysiloxane with the residual functional group in silicon gel and the functional group which reacts, and quantity must be chosen, or it is necessary to wash or wipe away after hardening. In the method of (3), since a silicone gel composition is an uncured state when heating, the ORGANO hydrogen polysiloxane given to the surface of this constituent is involved in by the convection into a silicone gel composition. As a result, a surface layer hardens unevenly, and a good protective layer is not obtained, and also control of the thickness of a surface protection layer and hardness is difficult. In order to harden the constituent which becomes a silicone elastomer layer or a silicone resin layer in the method of (4) after hardening a silicone gel composition, a process becomes complicated and, also in time, is disadvantageous. [0009]Then, SUBJECT of this invention is providing the manufacturing method which can manufacture easily the silicone complex which consists of base phase which consists of silicon gels, and a surface-protection phase which has this base phase and good adhesion and was made in a reliable silicone elastomer or silicone resin at an easy process.

[0010]

[Means for Solving the Problem]That is, this invention is a manufacturing method of a silicone complex which consists of a surface layer which consists of a silicone elastomer or silicone resin formed in base phase which consists of silicon gels, and its surface, [0011](a-1) It averages among all the organic groups combined with a silicon atom contained in one molecule. More than 0.025 mol % is an alkenyl group. Organopolysiloxane at least two sorts of whose organic groups except this alkenyl group are more than 10 mol %, respectively and. which has the viscosity at 25 ** in the range of 50 - 100,000 cP which has an average of 0.1-2 alkenyl groups combined with a silicon atom in one molecule, [0012](a-2) It is the ORGANO hydrogen polysiloxane which has the hydrogen atom combined with a silicon atom in [1-50] one molecule, per [which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (a-1) /] alkenyl group -- what is blended so that it may become 0.3-2.0 individual -- and [0013](a-3) On the surface of a process of providing a phase which consists of a hardenability silicone composition (A) which contains and hardens an addition reaction catalyst and turns into a gel hardened material, and this phase [0014](b-1) inside 0.05 [an average of 1 of all the organic groups combined with a silicon atom contained in one molecule -- more than mol % being an alkenyl group, and, And viscosity at 25 ** is in the range of 50 - 100,000 cP, and inside of all the organic groups combined with a silicon atom in said organopolysiloxane (a-1) -- more than 10 mol % -- an organic group to contain is not contained -- organopolysiloxane which has two or more alkenyl groups combined with a silicon atom in one molecule, [0015](b-2) It is a hydrogen atom combined with a silicon atom in one molecule 5-100 It is the ORGANO hydrogen polysiloxane which ****, per [which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (b-1) / 1 alkenyl group -- what is blended so that it may become 0.3-2.0 individual -- and [0016](b-3) A process of providing a layer which consists of a hardenability silicone composition containing an addition reaction catalyst which hardens and becomes the shape of an elastomer, or the shape of resin. And a manufacturing method which has the process of presenting hardening with a layer which consists of a phase which consists of said constituent (A), and a constituent (B) simultaneously is provided.

[0017]Silicon gel is specified to JIS K-6301 and means a silicone hardened material whose rubber hardness measured with A type spring-loaded hardness scale is 0. [as used herein] Gel means that a silicone hardened material is in a state of such silicon gel. Generally, this silicon gel has the three-dimensional network structure with a low degree of cross linking, and it changes with stress, and vibration is absorbed or it shows

mobility. On the other hand, rubber hardness of the shape of an elastomer measured with said A type springloaded hardness scale is large -100 from 0. It means having the hardness of the range of the following. With the shape of resin, rubber hardness measured with said A type spring-loaded hardness scale. It means having hardness of 100 or more.

[0018]A silicone composition (A) silicone composition (a) turns into a gel hardened material by hardening, including (a-1) - (a-3) an ingredient as an essential ingredient.

[0019](a-1) Organopolysiloxane (a-1) of organopolysiloxane **, It averages among all the organic groups combined with a silicon atom contained in one molecule. More than 0.025 mol % is an alkenyl group, An alkenyl group at least two sorts of whose organic groups except this alkenyl group are more than 10 mol %, respectively and, which has the viscosity at 25 ** in the range of 10 - 100,000 cP which was combined with a silicon atom in one molecule is averaged. It has 0.1-2 pieces. Silicon gel will be hard to be obtained if there are few rates of an alkenyl group among all the organic groups combined with a silicon atom contained in one molecule than average 0.025-mol %. As this alkenyl group, a vinyl group, an allyl group, an isopropenyl group, a cyclohexenyl group, etc. are mentioned. (a-1) Since it reacts easily according to that composition of an ingredient is easy, and many kinds of catalysts, a vinyl group is preferred.

group, a propyl group, a butyl group, a pentyl group, a hexyl group, Aralkyl groups, such as alkyl groups, such as an octyl group and a decyl group, benzyl, and 2-phenylethyl group, Aryl groups, such as a phenyl group, a tolyl group, and a naphthyl group, or a chloromethyl group which replaced some or all of a hydrogen atom of these bases with a halogen atom etc., 1-10 carbon atoms, such as a 3,3,3-trifluoropropyl group, -- a monovalent hydrocarbon radical of unsubstituted [with 1-8 carbon atoms] or substitution is mentioned preferably. From that composition is easy and the heat resistance of base phase which consists of silicon gel obtained, and a physical property being excellent. although a methyl group, a phenyl group, and a 3,3,3-trifluoropropyl group are preferred, in (b-1) ingredient mentioned later and an ingredient (a-1) from a point which is immiscible nature, at least two kinds in these organic groups receive all the organic groups -- more than each 10 mol % -- to contain is needed.

[0021]viscosity at 25 ** of this organopolysiloxane (a-1) -- 50 - 100,000 cP -- it is 100 - 10,000cP preferably. If viscosity is lower than 50cP, since it is easy to flow through a silicone composition (A) obtained, workability will fall, and physical properties, such as a loss tangent etc. of a hardened material produced by hardening this constituent, will become dissatisfied. The workability of a constituent obtained on the other hand even if viscosity is higher than 100,000 cP worsens.

[0022]In order for a silicone composition (A) to turn into a gel hardened material by hardening, (a-1) Average among all the organic groups combined with a silicon atom which contains an ingredient in one molecule It is required to be what more than 0.025 mol % is an alkenyl group, and has 0.1-2 average alkenyl groups combined with a silicon atom in one molecule. Straight chain shape or branched state may be sufficient as molecular structure of this organopolysiloxane (a-1). A form of these mixtures may be sufficient. [0023]What is expressed with the following general formula as organopolysiloxane of (a-1) ingredient which

was stated above, for example is mentioned.

[Formula 1]

$$\begin{pmatrix} \mathsf{CH_3} & \mathsf{CH_3} \\ \mathsf{CH_3} & \mathsf{SiO} \\ \mathsf{CH_3} & \mathsf{CH_3} \end{pmatrix}_{\mathsf{L}} \begin{pmatrix} \mathsf{CH_3} \\ \mathsf{SiO} \\ \mathsf{CH_3} \\ \mathsf{CH_3} \end{pmatrix}_{\mathsf{L}} \begin{pmatrix} \mathsf{CH_3} \\ \mathsf{SiO} \\ \mathsf{CH_3} \\ \mathsf{CH_3} \end{pmatrix}_{\mathsf{L}}$$

[Here x, and y and z, $0 \le x \le 1.9$, As opposed to all the organicity which positive number;I of $0.1 \le y \le 2$ and x+y=2, m, and n are 0 or a positive integer, respectively, and at least two sorts in;CH₃-, C_6H_5 -, and $CF_3C_2H_4$ -combined with the silicon atom, respectively. It is suitably chosen so that it may become more than 10 mol %.] [0024](a-2) An ORUGANOHAIDOROJIEN PORISHIROKISANORUGANO hydrogen polysiloxane (a-2), It must react to an alkenyl group in the aforementioned (a-1) ingredient, silicon gel must be formed, and a hydrogen atom combined with a silicon atom must exist in [at least one] one molecule. Such a hydrogen atom may be combined with an end of the ORGANO hydrogen polysiloxane chain, or which an intermediate silicon atom. [0025]As univalent atoms or bases other than a hydrogen atom combined with a silicon atom of the ORGANO hydrogen polysiloxane (a-2), What was illustrated as bases other than an alkenyl group combined with a silicon atom of organopolysiloxane of the aforementioned (a-1), and same thing can be illustrated. (a-2) Since an ingredient's being easily compoundable and the heat resistance of silicon gel obtained become the more outstanding thing, a methyl group is preferred.

[0026](a-2) hydrogen atom [] combined with a silicon atom in (a-2) ingredient to one alkenyl group combined with loadings of an ingredient, and a silicon atom in an ingredient (a-1) -- 0.3-2.0 individual -- desirable -- -- it is the quantity used as 0.5-1.5 individual. The heat resistance of base phase will worsen by an unreacted alkenyl group which remains in silicon gel obtained as it is a quantity smaller than 0.3 piece. There is a possibility of foaming in the case of hardening of a constituent (A) by their being more quantity than 2.0 pieces. [0027](a-2) as [mention / the ingredient needs to have (a-1) ingredient and compatibility, and / them / ingredient / later / for that purpose] -- the organic groups with a main ingredient (a-2) need to choose so that highly [an organic group (for example, methyl group) and compatibility which an ingredient (a-1) mainly has]. Or in order for a constituent (A) to become gel by hardening, the ingredient (a-2) needs to have the hydrogen atom combined with a silicon atom in [1-50] one molecule. Although viscosity in particular is not restricted,

[0028]As an ORGANO hydrogen polysiloxane of (a-2) ingredient which was stated above, what is expressed with the following general formula is mentioned.

since easy to compound an ingredient (a-2) and workability are good, in 25 **, it is preferred that it is the range

[Formula 2]
$$\begin{pmatrix} CH_3 \\ CH_3 - Si0 \end{pmatrix} + \begin{pmatrix} CH_3 \\ I \\ Si0 \end{pmatrix} + \begin{pmatrix} I \\ I \\ Si0 \end{pmatrix} + \begin{pmatrix} CH_3 \\ I \\ Si-H \end{pmatrix}$$

of 10 - 1,000 cP.

[Here, x and y are the positive numbers of x>=0, y>=0, and x+y=2, and;m and n are the integers of $0 \le x \le 0$, $0 \le x \le 0$, and $0 \le x \le 0$.]

[0029](a-3) What kind of catalyst known as what promotes an addition reaction (hydrosilylation reaction) of an alkenyl group combined with a silicon atom in an addition reaction catalyst addition reaction catalyst (a-3) and an ingredient (a-1) and a hydrogen atom combined with a silicon atom in an ingredient (a-2) may be sufficient. It is used by platinum metal system catalyst and Usually, for example, chloroplatinic acid, denaturing alcohol

chloroplatinic acid, Platinum system catalysts, such as a complex of chloroplatinic acid and a vinyl siloxane, and a chloroplatinic acid-2-ethylhexanol solution, and tetrakis (triphenyl phosphine) Palladium system catalysts, such as a mixture of palladium, palladium black, and triphenyl phosphine, a rhodium catalyst, etc. are mentioned. A chloroplatinic acid-2-ethylhexanol solution is especially preferred. [0030]What is called a catalyst amount may be sufficient as loadings of these catalysts. usually, the total

quantity of an ingredient (a-1) and (a-2) ingredient is received -- it is the range of 0.1 - 100 ppm (catalyst

metallic element conversion). [0031]A silicone composition (B) silicone composition (B) becomes a surface layer of the shape of an elastomer, or the shape of resin by hardening, including (b-1) - (b-3) an ingredient as an essential ingredient. [0032](b-1) Organopolysiloxane (b-1) of organopolysiloxane **, More than average 0.05 mol % is an alkenyl group among all the organic groups combined with a silicon atom contained in one molecule, And viscosity at 25 ** is in the range of 10 - 100,000 cP, and more than 10 mol % among all the organic groups combined with a silicon atom in said organopolysiloxane (a-1) -- at least a kind of organic group to contain is not contained -- it has two or more alkenyl groups combined with a cay original child in one molecule. When there are few rates of an alkenyl group among all the organic groups combined with a silicon atom contained in one molecule than an average of 0.05-mol %, a hardened material of the shape of Hellas Thau or the shape of resin is difficult to get. A thing of illustration about an ingredient (a-1) as this alkenyl group is mentioned, and a vinyl group is preferred.

[0033]Although what was illustrated about an ingredient (a-1) also as organic groups other than an alkenyl group combined with a silicon atom, and same thing are mentioned and it is a methyl group and a phenyl group preferably, as opposed to all the organic groups combined with a silicon atom in an ingredient (a-1) in (b-1) ingredient from a point which are (a-1) ingredient mentioned above and immiscible nature -- more than 10 mol % -- a thing of the organic groups to contain for which a kind is not contained at least is required. [0034](b-1) viscosity at 25 ** of organopolysiloxane of an ingredient -- 50 - 100,000 cP -- it is 100 - 10,000 cP preferably. If viscosity is lower than 50 cP, it will be easy to flow through a silicone composition (b) obtained, and a physical property of a hardened material produced by hardening this constituent will become dissatisfied. On the other hand, if viscosity is higher than 100,000 cP, the workability of a constituent obtained will worsen. [0035]In order for a silicone composition (B) to turn into a hardened material of the shape of an elastomer, or

the shape of resin by hardening, (b-1) an average of 0.05 among all the organic groups combined with a silicon atom which contains an ingredient in one molecule -- more than mol % is an alkenyl group, and it is necessary to have two or more alkenyl groups combined with a silicon atom in one molecule [0036]It is necessary to have immiscible nature to this (b-1) ingredient and an ingredient (a-1). It is dependent on a kind of a kind of organic groups other than an alkenyl group combined with a silicon atom in (b-1) ingredient as mentioned above, quantity, and (a-1) ingredient whether it has compatibility or it is immiscible nature. When a grade of compatibility is described about a typical organic group, low-grade alkyl groups (it is called the 1th group for convenience), such as a methyl group and an ethyl group, have high compatibility to mutual, and, generally the organopolysiloxane which an organic group becomes mainly from these has compatibility, on the other hand, bases, such as a phenyl group, a 3,3,3-trifluoropropyl group, and a ** polyether group (the -- it is called II group), since an aforementioned basis and compatibility of the Ith group are low, organopolysiloxane which mainly consists of an organic group of the Ith group - mainly -- the --

organopolysiloxane which has an organic group of II group does not have compatibility. Being still more concrete and the following as an example an ingredient (b-1) indicates immiscible nature to be to (a-1) ingredient are mentioned.

[0037](a-1) Ingredient: example 1) Organic groups other than an alkenyl group combined with a silicon atom are a methyl group and a 3,3,3-trifluoropropyl group, Organopolysiloxane, an ingredient (b-1) whose rate of the latter 3,3,3-trifluoropropyl group is more than 30 mol % on an average: Poly dimethylsiloxane.

[0038](a-1) example 2) Ingredient: organopolysiloxane, ingredient (b-1):poly dimethylsiloxane whose rate of the latter phenyl group organic groups other than an alkenyl group combined with a silicon atom are a methyl group and a phenyl group, and is more than 10 mol % on an average.

[0039](a-1) example 3) Organic groups other than an alkenyl group combined with a silicon atom are a methyl group and a 3,3,3-trifluoropropyl group, Organopolysiloxane whose rate of the latter 3,3,3-trifluoropropyl group is more than 30 mol % on an average, (b-1) Organopolysiloxane whose rate of the latter phenyl group organic groups other than an alkenyl group combined with a silicon atom are a methyl group and a phenyl group, and is more than 10 mol % on an average.

[0040](b-1) Straight chain shape or branched state may be sufficient as molecular structure of organopolysiloxane of an ingredient. A form of these mixtures may be sufficient. As organopolysiloxane of (b-1) ingredient which was stated above, what is expressed with the following general formula is mentioned. Formula 31

[Here, I, m, n, and p are 0 or a positive integer, respectively, and at least one of I, m, and n is 0.] [0041](b-2) The ORGANO hydrogen polysiloxane (b-2) used for ORGANO hydrogen polysiloxane this invention reacts to the alkenyl group in the aforementioned (b-1) ingredient, and forms the hardened material of the shape of an elastomer, or the shape of resin. Therefore, the hydrogen atom combined with a silicon atom must exist in [at least one] one molecule. Such a hydrogen atom may be combined with the end of the ORGANO hydrogen polysiloxane chain, or which intermediate silicon atom.

[0042]What was illustrated as bases other than the alkenyl group combined with the silicon atom of organopolysiloxane of the aforementioned (b-1) as bases other than the hydrogen atom combined with the silicon atom of the ORGANO hydrogen polysiloxane (b-2), and the same thing can be illustrated. (b-2) A methyl group is preferred at that an ingredient is easily compoundable and the point that the heat resistance of silicon gel obtained becomes the more outstanding thing.

[0043](b-2) hydrogen atom [] combined with a silicon atom in (b-2) ingredient to one alkenyl group combined with loadings of an ingredient, and a silicon atom in an ingredient (b-1) — 0.3-2.0 individual — desirable — — it is the quantity used as 0.5-1.5 individual. Heat resistance which this layer has will worsen by an alkenyl group which remains in a surface layer obtained as it is a quantity smaller than 0.3 piece. There is a possibility of foaming in the case of hardening of a silicone composition (B) obtained as them are more quantity than 2.0 pieces.

[0044]In order for this (b-2) ingredient to have compatibility to the aforementioned (b-1) ingredient, organic groups which an ingredient (b-2) has need to be main organic groups which (b-1) ingredient has, and a high

organic group of compatibility. What is necessary is just to choose the suitable ORGANO hydrogen polysiloxane in the light of a relation of compatibility mentioned above.

[0045]It is a hydrogen atom which combined an ingredient (b-2) with a silicon atom in order for a hardened material produced by making harden a silicone composition (b) to become a thing of the shape of an elastomer, or the shape of resin in one molecule It is necessary to ****. 5-100 Although viscosity in particular is not restricted, since easy to compound an ingredient (b-2) and workability are good, in 25 **, it is preferred that it is the range of 10 - 1,000 cP.

[0046]As an ORGANO hydrogen polysiloxane of (b-2) ingredient which was stated above, what is expressed with the following general formula is mentioned.

[Here, number;m and n of x>=0, y>=0, and x+y=2 of x and y are 0<=m and an integer of 3<=n<=100.] [0047](b-3) As an addition reaction catalyst used for an addition reaction catalyst (b-3) ingredient, the same catalyst as what was explained as an ingredient (a-3) can be used. What is called a catalyst amount may be sufficient as loadings. the total quantity with (b-2) is received with usually (b-1) — it is the range of 0.1 - 100 ppm (catalyst metallic element conversion).

[0048]It is possible by changing the presentation of a constituent (B), and the amount of the constituent (B) used to adjust the hardness and thickness of a surface layer of a complex which are obtained. Thickness of a surface layer can be thickened by that it is concrete and being able to raise the hardness of a hardened material and increasing the amount (injection rate) of the constituent (B) used by increasing the quantity of the alkenyl group in an ingredient (b-1), and the quantity of the silicon atom absorbed water matter atom (Si-H group) in an ingredient (b-2).

[0049]In one desirable mode of this invention, a self adhesiveness silicone composition is used for a constituent (B). The self adhesiveness of a constituent (B) makes a bonding assistant the ORGANO siloxane etc. which have in a molecule the alkenyl group which combined the hydrogen atom combined with alkoxy silyl groups and a silicon atom with the ORGANO siloxane or alkoxy silyl groups which it has in a molecule, and a silicon atom, It can be reached by blending into a constituent.

[0050]In other combination drug silicone compositions (A) and silicone compositions (B), an additive agent in which adding conventionally to a hardenability silicone composition besides the aforementioned ingredient is known may be added further if needed. For example, they are colorant, such as flame retarders, such as reaction controlling agents, such as an acetylenealcohol compound, manganese carbonate, and carbon black, a color, and paints, heat-resistant stabilizer, oilproof stabilizer, etc. In order to raise the vibration absorption of a hardened material produced by hardening, an organic polymer bulking agent containing a low boiling compound may be blended with an inside if needed. however, a hardened material obtained when it is that a hardened material produced by making harden these arbitrary additive agents in the case of a constituent (A) serves as gel, and a constituent (B) — the shape of an elastomer, or resin — ** — things are not checked.

[0051]In order to provide a phase which consists of a process silicone composition (A) which provides a phase which consists of silicone compositions (A), there is [which is poured / which is sprayed / which applies this

constituent (A) / on the inside of a mold of predetermined shape made into the purpose, or a support surface / in 1 a method of making it extruding [to eiect 1, being dropped, fall, etc.

[0052]It applies [which applies a silicone composition (B)] to the surface of a phase which consists of a silicone composition (A) provided in the process above which provides a layer which consists of silicone compositions (B) by making it like by pouring [to spray] in, ejecting, extruding, etc., and a surface layer of a constituent (B) is provided in it. Since each main ingredients are immiscible nature, a constituent (A) and a constituent (B) are immiscible nature also as a constituent. Therefore, by having laminated, it does not mix mutually, or these constituents are diffused and do not suit.

[0053]Although that by which an unhardened surface layer was laminated by unhardened base phase by a process of the <u>curing process</u> above is obtained, curing treatment is simultaneously presented with these as one. A silicone complex which consists of a surface layer which consists of base phase, an elastomer, or resin which consists of silicon gels by this can be obtained. As conditions for hardening, the usual curing conditions may be sufficient, for example, it is the temperature about 60 - 150 **, and is 30-180. Heat-treatment about a part may be sufficient.

[0054]Since a layer of a silicone elastomer or resin whose internal silicon gel phase is a surface layer as a stress relaxation phase functions effectively as a surface protection layer, a silicone complex obtained by a method of <u>use</u> this invention is useful as coating of various kinds of electrical and electric equipment and the surface of electronic parts or a semiconductor device. A silicone complex of this invention can be fabricated on any surface of substrates, such as various kinds of plastics, metal, and glass.

[0055]

[Example]In the following statements, viscosity shows the value at 25 **.

It is Example 1 (1) average and is the following formula. : [Formula 5]

$$\begin{array}{c} \text{CH}_3 \\ \text{CH}_2 = \text{CH} - \overset{\bullet}{\text{Sio}} \\ \overset{\bullet}{\text{I}} \\ \overset{\bullet}{\text{CH}}_3 \\ \overset{\bullet}{\text{CH}}_3 \end{array} \begin{array}{c} \overset{\bullet}{\text{CH}}_3 \\ \overset{\bullet}{\text{Sio}} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \end{array} \begin{array}{c} \overset{\bullet}{\text{CH}}_3 \\ \overset{\bullet}{\text{Sio}} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \end{array} \begin{array}{c} \overset{\bullet}{\text{CH}}_3 \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \end{array} \begin{array}{c} \overset{\bullet}{\text{C}}_{\text{H}_3} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \\ \overset{\bullet}{\text{C}}_{\text{H}_3} \end{array} \begin{array}{c} \overset{\bullet}{\text{C}}_{\text{H}_3} \\ \overset{\bullet}{\text{C}_$$

It is expressed [(the constitutional unit is arranged at random among the formula), and], and is viscosity. Organopolysiloxane 100 weight section of 700cP, the following formula: [Formula 6]

$$C_{\bullet}H_{\delta}-Si \leftarrow \begin{pmatrix} CH_{\delta} \\ + \\ OSi-H \\ - \\ CH_{\delta} \end{pmatrix}_{\delta}$$

[0056]It came out, and it was expressed, and ORGANO hydrogen polysiloxane 3.8 weight section which is viscosity 2cP, and 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (A-1) was obtained.

[0057](2) It is an average and is the following formula. : [Formula 7]

It is come out and expressed, is organopolysiloxane 100 weight section and average which are viscosity

100cP, and is the following formula. : [Formula 8]

[0058]ORGANO hydrogen polysiloxane 7 weight section which it is expressed [(the constitutional unit is arranged at random among the formula) and], and is viscosity 10cP, And 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (B-1) was obtained.

[0059](3) 20 g of silicone compositions (A-1) were poured into an aluminum petri dish 5 cm in diameter, 2 g of silicone compositions (B-1) were continuously laminated on the surface, and laminated material was obtained. When this laminated material was heated at the temperature of 60 ** for 2 hours, the transparent silicone complex which consists of a surface layer of the shape of an elastomer without adhesiveness and gel base phase which has cold resistance was obtained.

[0060] Example 2 (1) The following structural formula : [Formula 9]

It is come out and expressed, is organopolysiloxane 100 weight section and average which are viscosity 1000cP, and is the following formula: [Formula 10]

$$\begin{array}{c} \text{CH}_3 \\ \text{CH}_3 - \text{SiO} \\ \text{CH}_3 - \text{SiO} \\ \text{CH}_3 \\ \text{CH}_3 \\ \end{array} \\ \begin{array}{c} \text{H} \\ \text{SiO} \\ \text{SiO} \\ \text{CH}_3 \\ \text{SiO} \\ \text{CH}_3 \\ \\ \text{CH}_4 \\ \\ \text{CH}_3 \\ \\ \text{CH}_3 \\ \\ \text{CH}_4 \\ \\ \text{CH}_5 \\$$

[0061]It is expressed [(the constitutional unit is arranged at random among the formula), and], and as ORGANO hydrogen polysiloxane 5 weight section and 5 ppm of chloroplatinic acid-2-ethylhexanol solutions which are viscosity 28cP, and (platinum conversion) a bulking agent, Crystalline silica powder 50 weight section was taught to the mixer, it mixed uniformly, and the silicone composition (A-2) was obtained.

[0062](2) It is an average and is the following formula. : [Formula 11]

$$\begin{array}{c} \text{CH}_2\text{=CH-SiO} \left(\begin{array}{c} \text{SiO} \\ \text{CH}_3 \end{array}\right) \\ \begin{array}{c} \text{CH}_3 \end{array}\right) \\ \begin{array}{c} \text{C}_{\text{B}} \\ \text{C}_{\text{B}} \\ \text{J} \end{array}\right) \\ \begin{array}{c} \text{C}_{\text{B}} \\ \text{C}_{\text{B}} \\ \text{J} \end{array}\right) \\ \begin{array}{c} \text{C}_{\text{B}} \\ \text{C$$

It is expressed [(the constitutional unit is arranged at random among the formula), and], and is viscosity. Organopolysiloxane 100 weight section of 700cP, the following structural formula : [Formula 12]

$$C_6H_8 - Si \leftarrow \begin{pmatrix} CH_3 \\ I \\ OSi-H \\ CH_3 \end{pmatrix}_s$$

[0063]It was come out and expressed, and ORGANO hydrogen polysiloxane 7 weight section of viscosity 2cp

and 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (B-2) was obtained. The 20 g aforementioned silicone composition (A-2) was poured into an aluminum petri dish 5 cm in diameter, and the 2 g silicone composition (B-2) was continuously laminated on the surface. When the obtained lamination constituent was heated at the temperature of 60 ** for 2 hours, the transparent silicone complex which consists of a surface layer of the shape of an elastomer without adhesiveness and a gel internal layer which has cold resistance was obtained. [0064]It is Example 3 (1) average and is the following formula: [Formula 13]

It is expressed [(the constitutional unit is arranged at random among the formula), and], and is viscosity. Organopolysiloxane 100 weight section of 800cP, the following structural formula: [Formula 14]

[0065]It was come out and expressed, and ORGANO hydrogen polysiloxane 15 weight section of viscosity 150cP and 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (A-3) was obtained.

[0066](2) The following structural formula: [Formula 15]

It is come out and expressed, is organopolysiloxane 100 weight section of viscosity 100cP, and an average, and is the following formula.: [Formula 16]

[0067]It is expressed [(the constitutional unit is arranged at random among the formula), and], and ORGANO hydrogen polysiloxane 8 weight section of viscosity 10cP, And 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (B-3) was obtained.

[0068](3) The 20 g silicone composition (A-3) was poured into an aluminum petri dish 5 cm in diameter, and the 2 g silicone composition (B-3) was continuously laminated on the surface. When the obtained laminated material was heated at the temperature of 60 ** for 2 hours, the transparent silicone complex which consists of a surface layer of the shape of an elastomer without adhesiveness and a gel internal layer which has cold resistance was obtained.

[0069]Example 4 (1) The following structural formula: [Formula 17]

It is come out and expressed, is organopolysiloxane 100 weight section of viscosity 1000cP, and an average, and is the following formula.: [Formula 18]

[0070]It is expressed [(the constitutional unit is arranged at random among the formula), and], and as ORGANO hydrogen polysiloxane 5 weight section, 5 ppm of chloroplatinic acid-2-ethylhexanol solutions, and (platinum conversion) the bulking agent of viscosity 100cP, Crystalline silica powder 50 weight section was taught to the mixer, it mixed uniformly, and the silicone composition (A-4) was obtained.

[0071](2) It is an average and is the following formula.: [Formula 19]

It is expressed [(the constitutional unit is arranged at random among the formula), and], and is viscosity. Organopolysiloxane 100 weight section of 800cP, the following structural formula: [Formula 20]

[0072]It came out, and it was expressed, and ORGANO hydrogen polysiloxane 30 weight section which is viscosity 150cP, and 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (B-4) was obtained.

[0073](3) The 20 g silicone composition (A-4) was poured into an aluminum petri dish 5 cm in diameter, and the 2 g silicone composition (B-4) was continuously laminated on the surface. When the obtained laminated material was heated at the temperature of 60 ** for 2 hours, the transparent silicone complex which consists of a surface layer of the shape of an elastomer without adhesiveness and a gel internal layer which has cold resistance was obtained

[0074]

[Effect of the Invention]According to this invention, it has high hardness, high intensity, high solvent resistance, etc. relatively, and the complex which consists of a surface protection layer with easy regulation of hardness and thickness and a stress relaxation layer which has low hardness, low intensity, high solvent resistance, adhesiveness, etc. relatively can be manufactured by one curing treatment.

* NOTICES *

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- 1. This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.**** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

a silicone elastomer or silicone resin formed in base phase which consists of silicon gels, and its surface, (a-1) Average among all the organic groups combined with a silicon atom contained in one molecule. More than 0.025 mol % is an alkenyl group, At least two sorts of organic groups except this alkenyl group are more than 10 mol %, respectively, . And viscosity at 25 ** is in the range of 50 - 100,000 cP. An alkenyl group combined with a silicon atom is averaged in one molecule. Organopolysiloxane which it has 0.1-2 pieces, (a-2) It is the ORGANO hydrogen polysiloxane which has the hydrogen atom combined with a silicon atom in [1-50] one molecule, Per [which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (a-1)] alkenyl group 0.3-2.0 What is blended so that it may become an individual, On and (a-3) the surface of a process of providing a phase which consists of a hardenability silicone composition (A) which contains and hardens an addition reaction catalyst and turns into a gel hardened material, and this phase. (b-1) inside 0.05 [an average of] of all the organic groups combined with a silicon atom contained in

[Claim 1]It is a manufacturing method of a silicone complex which consists of a surface layer which consists of

phase. (b-1) inside 0.05 [an average of] of all the organic groups combined with a silicon atom contained in one molecule -- more than mol % being an alkenyl group, and, And viscosity at 25 ** is in the range of 50 - 100,000 cP, and inside of all the organic groups combined with a silicon atom in said organopolysiloxane (a-1) -- more than 10 mol % -- an organic group to contain is not contained. Organopolysiloxane which has two or more alkenyl groups combined with a silicon atom in one molecule, (b-2) It is a hydrogen atom combined with a silicon atom in one molecule 5-100 It is the ORGANO hydrogen polysiloxane which *****, Per [which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (b-1)] alkenyl group 0.3-2.0 What is blended so that it may become an individual, And (b-3) a manufacturing method which has the process of presenting hardening with a layer which consists of a process of providing a layer which consists of a hardenability silicone composition containing an addition reaction catalyst which hardens and becomes the shape of an elastomer, or the shape of resin, a phase which consists of said constituent (A), and a constituent (B) simultaneously.

[Translation done.]